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Nuclear isomerism in odd lead isotopes

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With 15 figures in the text

Introduction

According to the nuclear shell model, isomeric transitions of the M4 type are expected to be found in nuclides of odd mass number when the number of protons or neutrons is just below a magic number. Regularities within these groups near the magic numbers 50 ($g_{9/2}-p_{1/2}$ transitions), 82 ($h_{11/2}-d_{3/2}$) and 126 ($i_{13/2}-f_{5/2}$) have been discussed by several authors, for example Hill [1, 2] and Mihelich and de-Shalit [3]. Similar regularities would be expected to occur in the neutron-deficient isotopes of lead, particularly in view of the closed shell of 82 protons which occurs in this case. Complementary programmes of investigating these isotopes with particular reference to the occurrence of isomers have been undertaken at the Nobel Institute of Physics and the Gustaf Werner Institute for Nuclear Chemistry.

Prior to the commencement of this work the only well-known lead isomers were Pb^{207m}, Pb^{204m} and Pb^{202m}. In addition, Hopkins [4] had reported activities of 5.6 sec ($E_\gamma = 0.89$ MeV) and 50 sec ($E_\gamma = 0.25$, 0.42 and 0.67 MeV), which he ascribed to Pb^{202m} and Pb^{201m} respectively. After the discovery of the 3.7-hr Pb^{202m} [5], the mass assignment of the 5.6-sec activity seemed very doubtful. Preliminary measurements, reported from this laboratory [6], had shown that Pb^{203m} had a half-life of the order of 10 sec, and had associated with it a gamma ray of about 0.9 MeV, and possibly also a lower energy gamma ray. During the course of the present investigations, Fischer [7] has published results which are in agreement with some of those presented here. She has observed the isomers Pb^{201m} and Pb^{203m} produced by Tl(p, 3n) reactions and finds the following half-lives and transition energies:

$$Pb^{201m}:\ T_{\frac{1}{2}} = 60.1 \pm 4.4 \text{ sec},\quad E_\gamma = 0.65 \pm 0.03 \text{ MeV}$$

$$Pb^{203m}:\ T_{\frac{1}{2}} = 6.73 \pm 0.44 \text{ sec},\quad E_\gamma = 0.86 \pm 0.04 \text{ MeV}.$$

The main purpose of the work reported here has been to see if any systematic trend existed in the half-lives and decay schemes of these lead isomers. From such information it would be possible to draw some conclusions about the variation of certain level spacings in this region, which is of particular interest in the light of shell theory. It should also be possible to determine the ground state spins of the lead isotopes. Further, it is of interest to compare the half-lives of such a closely related group of isomeric transitions with the theoretical predictions.

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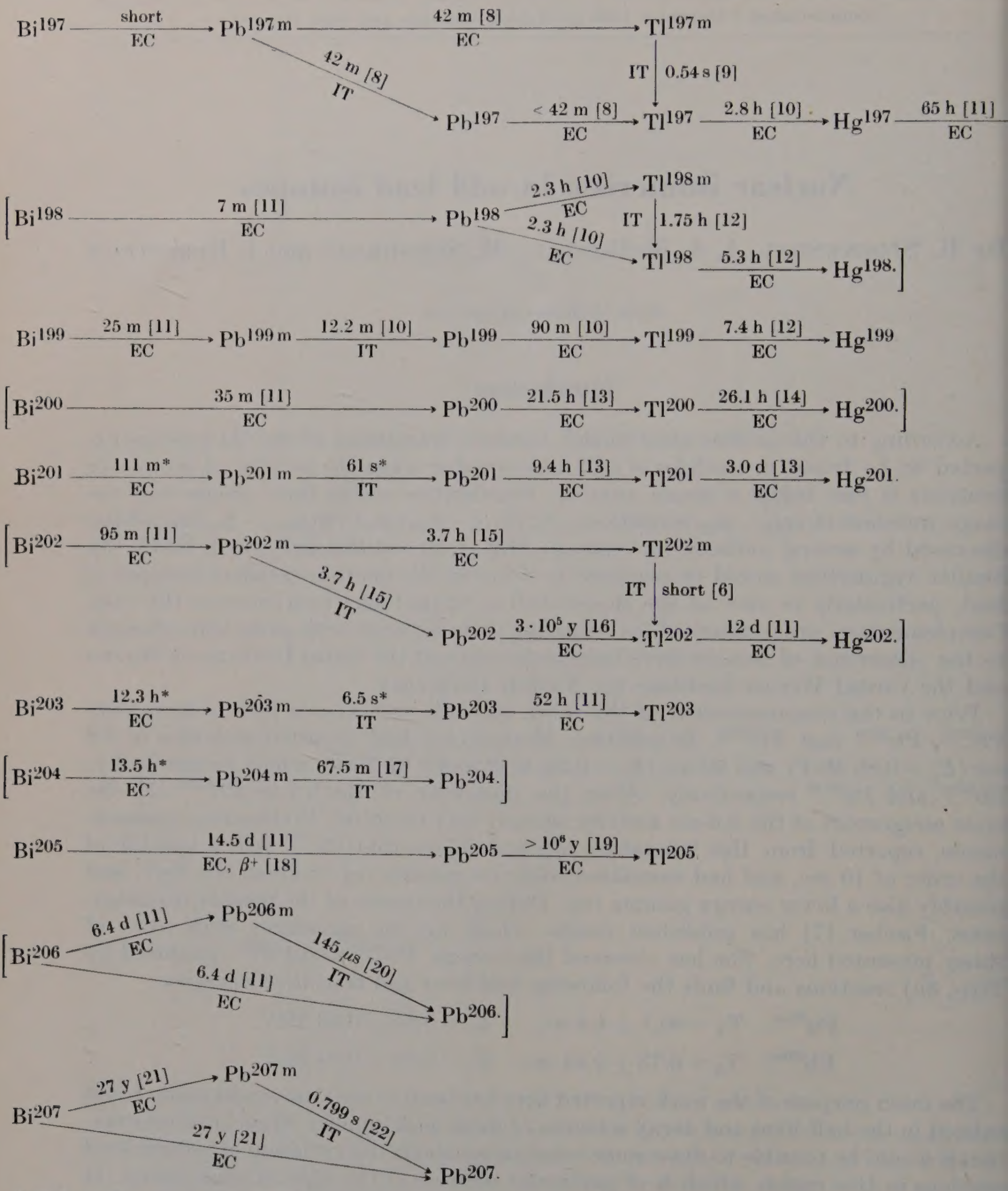
Bi197 short EC Pb197m 42 m [8] EC Tl197m
42 m [8] IT
IT 0.54 s [9]
Pb197 < 42 m [8] EC Tl197 2.8 h [10] EC Hg197 65 h [11] EC
[Bi198 7 m [11] EC Pb198 2.3 h [10] EC Tl198m
IT 1.75 h [12]
2.3 h [10] EC Tl198 5.3 h [12] EC Hg198.]
Bi199 25 m [11] EC Pb199m 12.2 m [10] IT Pb199 90 m [10] EC Tl199 7.4 h [12] EC Hg199
[Bi200 35 m [11] EC Pb200 21.5 h [13] EC Tl200 26.1 h [14] EC Hg200.]
Bi201 111 m* EC Pb201m 61 s* IT Pb201 9.4 h [13] EC Tl201 3.0 d [13] EC Hg201.
[Bi202 95 m [11] EC Pb202m 3.7 h [15] EC Tl202m
3.7 h [15] IT
IT short [6]
Pb202 3 · 10⁵ y [16] EC Tl202 12 d [11] EC Hg202.]
Bi203 12.3 h* EC Pb203m 6.5 s* IT Pb203 52 h [11] EC Tl203
[Bi204 13.5 h* EC Pb204m 67.5 m [17] IT Pb204.]
Bi205 14.5 d [11] EC, β⁺ [18] Pb205 > 10⁶ y [19] EC Tl205
[Bi206 6.4 d [11] EC Pb206m
145 μs [20] IT
6.4 d [11] EC Pb206.]
Bi207 27 y [21] EC Pb207m
0.799 s [22] IT
27 y [21] EC Pb207.

* This work.

The decay chains above, based on the most recent information, are of interest for our experiments (chains within brackets are of secondary interest). In the cases of Bi^{197}, Bi^{199}, Bi^{201}, Bi^{202}, Bi^{203} and Bi^{204} there are probably transitions, although not indicated, bypassing the isomeric states in lead.

Experimental procedures

a) Milking of short-lived lead activities

Targets of natural lead were bombarded with 25-MeV deuterons in the Nobel Institute cyclotron and with 40-, 50-, 70- and 100-MeV protons in the Uppsala synchrocyclotron. In each case the target was dissolved in nitric acid, and the bulk of the Pb precipitated by the addition of HCl in an ice bath and centrifuged off. The remaining solution containing the radioactive Bi and the rest of the Pb was then boiled under addition of more HCl until no traces of nitrate remained. After dilution to an approximate normality of 0.3, the solution was slowly passed through an ion-exchange column of Dowex 1 resin, 200 mesh, which was heated to the boiling point of iso-propyl alcohol (82.3°C). As has been shown by Kraus and Nelson [23], this results in a very effective separation of Pb and Bi, the Bi adhering strongly to the resin.

With the application of pressure by a syringe the column could be washed with $0.3N$ HCl and the eluate transferred to a scintillation spectrometer within three seconds of the commencement of the milking. The scintillation detector (a $1'' \times 1''$ diameter NaI(Tl) crystal coupled to a photo-multiplier) was used in conjunction with either a single-channel analyser or a 15-channel analyser. The resolution of this spectrometer was 8 % on the Cs^{137} calibration line. In this way it was possible to investigate the gamma spectrum of any lead activity washed through the column, and its half-life could also be measured. In cases in which the half-lives were of the order of seconds they were measured by photographing the scalers of the appropriate channel or channels with a movie camera.

b) β-spectrometer measurements

The Bi activity was washed off the column using $2N$ H_2SO_4, and, in order to remove the chloride, boiled until white SO_3 fumes appeared. It was then made up to be approximately $1N$ in H_2SO_4 and a few drops of hydrazine sulphate solution added. The Bi was then plated on to a Cu foil (~ 10 mg/cm^2) all of which except an area corresponding to the desired source shape (usually 2×15 mm) was masked off with nail polish. A Pt stirrer was used as an anode, and an applied potential of from 4 to 9 volts used. In most cases 50–60 % of the activity was transferred to the source, in times varying from half an hour to 4 hours.

The conversion-electron spectrum was studied in double-focusing spectrometers [24, 25] and in a permanent magnet β-spectrometer for photographic recording [26] (with the activity evaporated on to a 0.2-mm Cu wire).

Experimental results and discussion

Pb^{197m} and Pb^{199m}

Because the information about these isomers which is now available is relevant to our discussion we mention them here, although we have not investigated Pb^{197m} and

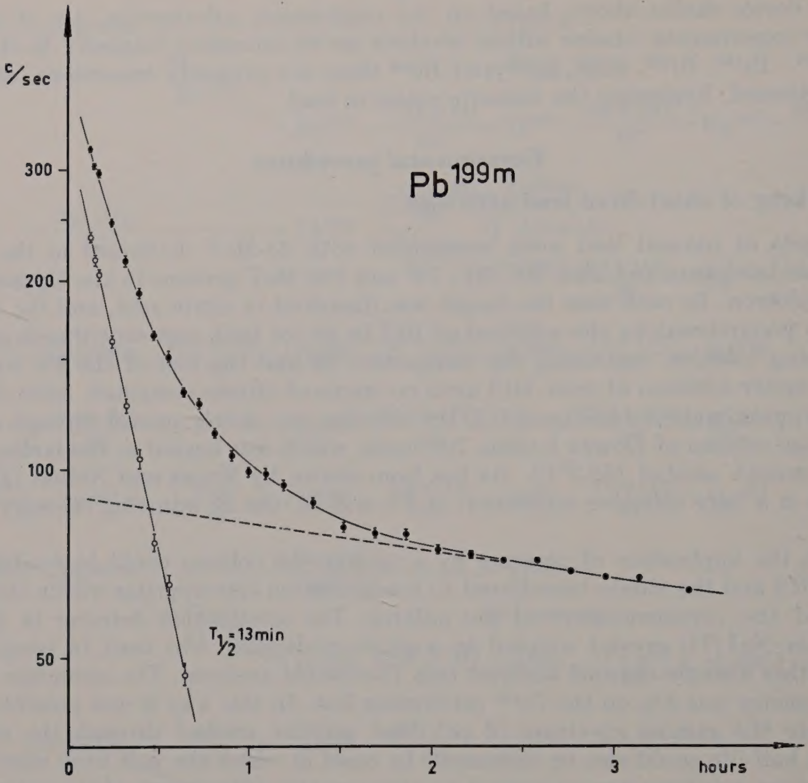

Fig. 1. Half-life of Pb^{199m} measured with a GM-counter.

have studied only the gamma spectrum of Pb^{199m}. The conversion spectra of these two isomers have been investigated at the Gustaf Werner Institute as part of the programme mentioned in the Introduction. Some preliminary results have been published [10], and we are grateful to Dr. Andersson and Dr. Arbman for permission to quote some additional information about these isomers, which they have produced by Tl(p, xn) reactions. The energies and half-lives concerned have been measured rather more accurately, and both isomeric transitions have been identified as M4 from the internal conversion data. The relevant results are included in Table 1.

Using the milking technique described above we have observed with a GM-counter a lead activity with a half-life of

$$T_{\frac{1}{2}\,(\mathrm{Pb}^{199m})} = 13 \pm 1 \text{ min}$$

as shown in Fig. 1. This activity was milked from radioactive bismuth produced by a 30-min bombardment of lead with 100-MeV protons. The scintillation spectrum of this activity was obtained from a milking taken from the column 18 minutes after the active bismuth had been adsorbed on it and washed with 0.3*N* HCl. Thus this milking was performed 138 minutes after the end of the bombardment. The spectrum was taken several times using a single-channel analyser (the multichannel analyser

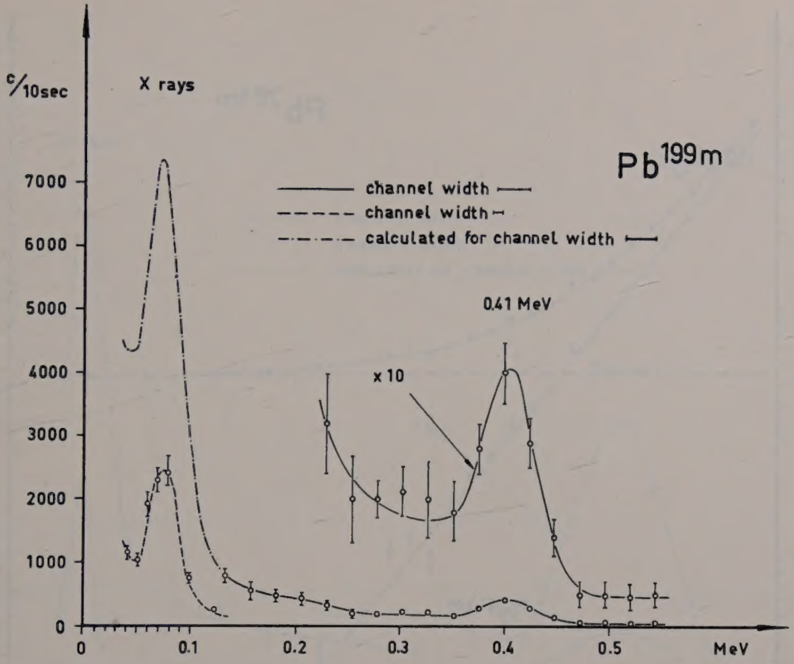

Fig. 2. Scintillation spectrum (corrected for background and decay) of 13-min Pb^{199m} recorded with a single-channel analyser. When the discriminator was set to accept pulses corresponding to energies above 550 keV, the counting rate was negligible. The low-energy part of the spectrum ($E < 130$ keV), taken with a smaller channel width, was transformed to a wide-channel spectrum by means of numerical integration.

was not available for these measurements). For different energies decay curves similar to that of Fig. 1 were plotted. From every such curve a straight line representing a 13-min activity was obtained by subtraction of the background, and extrapolation was made to time 0 (start of measurements). From the extrapolated values for different energies the spectrum of the 13-min activity was obtained. This is shown in Fig. 2. The energy of the observed photo peak is found to be

$$E_{\gamma(\mathrm{Pb}^{199m})} = 410 \pm 30 \text{ keV}.$$

It seems certain that this 410-keV photo peak is that of the 424-keV gamma ray of Pb^{199m} ($T_{\frac{1}{2}} = 12.2$ m), observed by Andersson and Arbman [8, 10], and this is now known to be an M4 transition. Considering the dependence of K conversion on the multipolarity and the variation of crystal efficiency with energy, one would expect any other gamma ray in cascade having a lower energy and presumably a lower multipolarity to exhibit a photo peak in the spectrum which would be many times as strong as that at 410 keV. Thus it is concluded from Fig. 2 that there is no other gamma ray having an energy greater than about 150 keV in cascade with the M4 transition.

If it is assumed that there is no other photo peak overlapping the X-ray peak, the K conversion coefficient

$$\alpha_K = 2.4 \pm 1.0$$

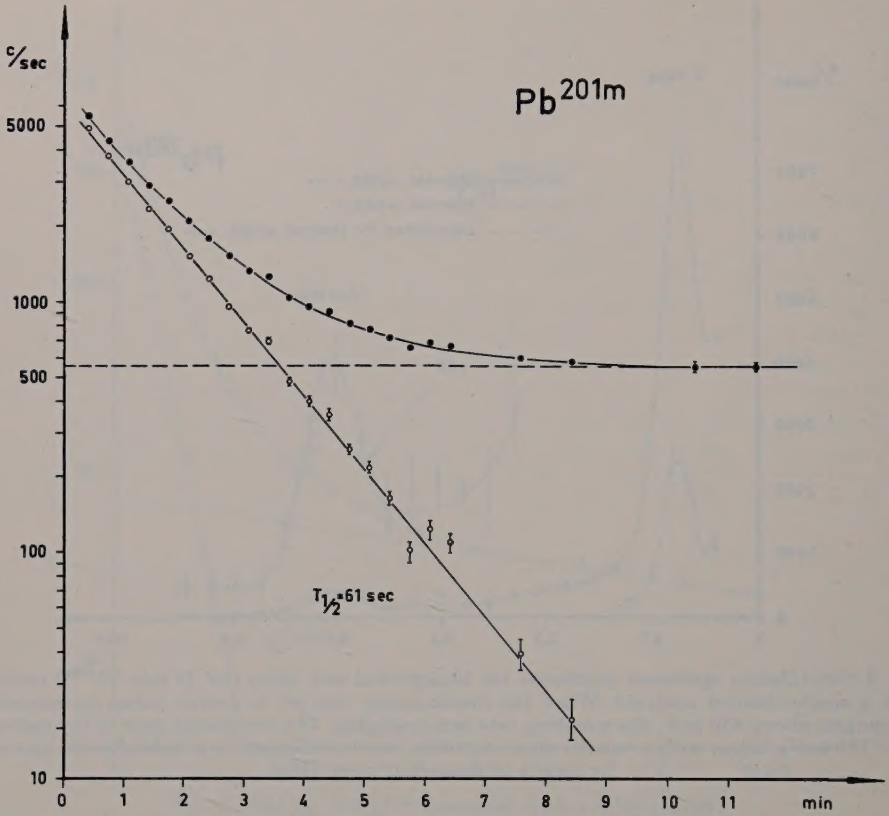

Fig. 3. Half-life of Pb^{201m} measured with a scintillation spectrometer. The discriminator setting was: 600 keV $< E <$ 650 keV.

can be calculated from the intensities (corrected for absorption in the glass (10 %) and crystal efficiency) of the X-ray peak and the photo peak. This is in agreement with the theoretical value of 2.39 for a pure M4 transition of this energy (0.20 for E4) [27]. Thus there should be no gamma ray with an energy between 40 and 150 keV overlapping the X-ray peak or the isomeric peak. If there exists any other gamma ray in the decay of Pb^{199m}, its energy must be less than about 40 keV. This tends to confirm that the isomeric transition of Pb^{199} goes to the ground state as was suggested by Andersson and Arbman [8].

Pb^{201m}

This isomer was observed following the bombardment of lead by 70-MeV protons. In the experiments using the milking technique, a lead activity with a half-life of

$$T_{\frac{1}{2}\,(Pb^{201m})} = 61 \pm 2 \text{ sec}$$

was found, this activity having associated with it a gamma ray of energy 620 ± 30 keV. Fig. 3 shows the decay of this activity for the case in which the discriminator of the

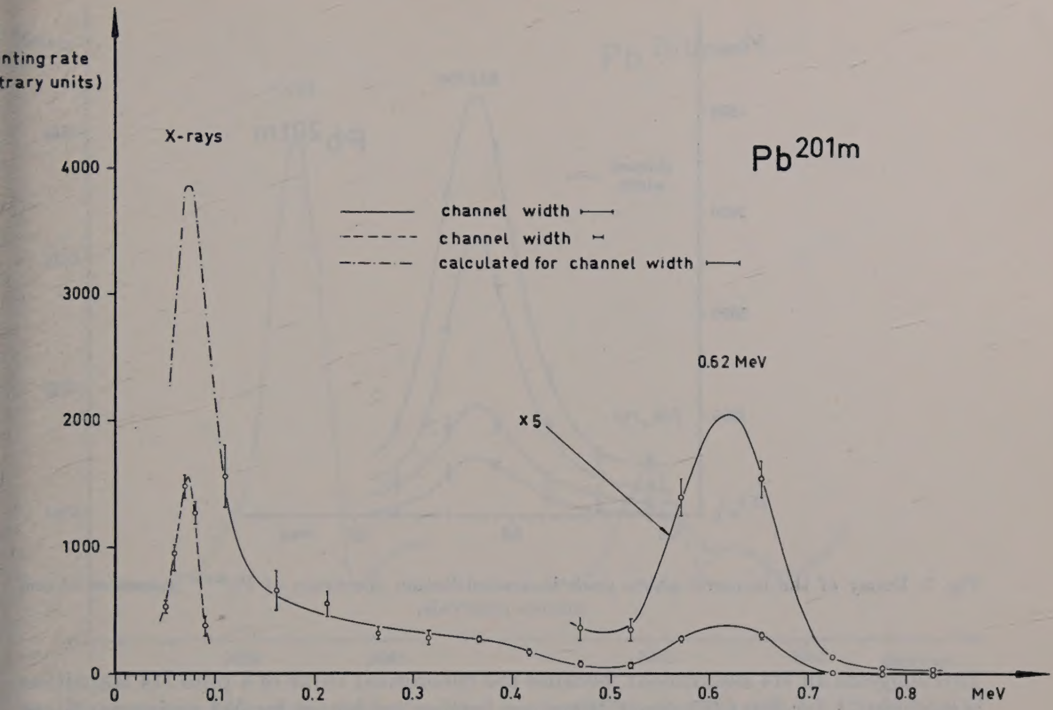

Fig. 4. Scintillation spectrum (corrected for background and decay) of 61-sec Pb^{201m} recorded with a single-channel analyser. With the discriminator set to accept pulses corresponding to energies above 860 keV, the counting rate was negligible. The spectrum was obtained by piecing together four different parts (from four different milkings) overlapping in energy. The low-energy part of the spectrum ($E < 110$ keV), taken with a smaller channel width, was transformed to a wide-channel spectrum by means of numerical integration.

scintillation spectrometer was set to accept pulses corresponding to energies between 600 keV and 650 keV. The background in Fig. 3 exhibits a decay which is of long half-life compared to the 61-sec activity.

The scintillation spectrum of the 61-sec activity (Fig. 4) was obtained in the same way as the Pb^{199m} spectrum (also here a single-channel analyser was used, as the multichannel analyser was not available). However, on account of the short half-life the whole spectrum could not be obtained from only one milking. From four milkings four different parts of the spectrum could be measured. Since these parts were always overlapping in energy, they could be pieced together to a spectrum covering the energy region above 40 keV. (Fig. 5 shows the decay of the isomeric photo peak obtained from another irradiation with the same bombarding energy). There is no indication of any other gamma ray above 150 keV. The K conversion coefficient, calculated from the intensities of the X-ray peak and the photo peak (absorption in the glass (10 %) and crystal efficiency taken into account) is

$$\alpha_K = 0.6 \pm 0.2.$$

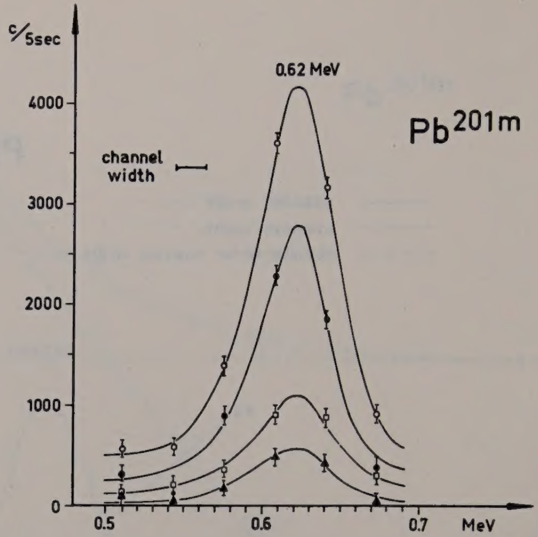

Fig. 5. Decay of the isomeric photo peak in a scintillation spectrum of Pb^{201m} measured at one minute intervals.

This suggests an M4 assignment, because the theoretical value of a pure M4 transition is 0.57 (0.071 for E4) [27]. Since there are further evidences for M4 assignment (see below), it seems certain that there is no gamma ray overlapping the X-ray peak or the isomeric peak. To sum up, therefore, we conclude that in addition to the isomeric gamma ray there is no gamma ray present with energy greater than about 40 keV in the decay of Pb^{201m}.

From another irradiation using the same bombarding energy, a source of the bismuth activity was prepared, as described above, for use in the double-focusing spectrometer of the Gustaf Werner Institute [24]. From its strong conversion lines (Fig. 6), measured with 0.3 % resolution, the energy of the isomeric gamma ray was found to be

$$E_{\gamma(\mathrm{Pb}^{201m})} = 629 \pm 1 \text{ keV},$$

and its K/L ratio

$$\mathrm{K/L} = 2.3 \pm 0.3.$$

From the energy difference between the K and L lines it was verified that, as expected, the conversion takes place in lead. The L lines were only partially resolved, but we were able to estimate

$$(\mathrm{L_I} + \mathrm{L_{II}})/\mathrm{L_{III}} = 4 \pm 1.$$

For a transition of this half-life and energy the calculations of Moszkowski [28] and the empirical analysis of Goldhaber and Sunyar [29] both indicate that the only possible assignments are M4 or E4. The conversion ratios for a 629-keV transition having either of these multipolarities are easily found from the tables of Rose and his collaborators [30], and are

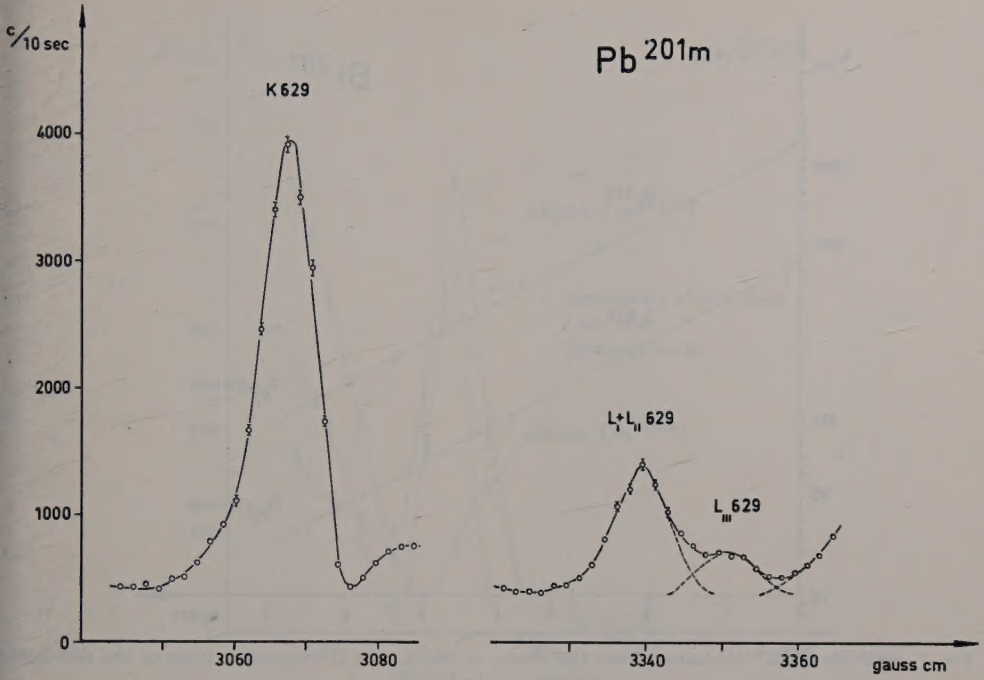

Fig. 6. Conversion lines of the 629-keV gamma ray in Pb^{201m} measured with 0.3 % resolution in a double-focusing β-spectrometer.

$$\text{M4:}\quad K/L = 2.7 \quad (L_I + L_{II})/L_{III} = 5.5$$
$$\text{E4:}\quad K/L = 1.3 \quad (L_I + L_{II})/L_{III} = 8.5.$$

Thus also the ratios of K/L and $(L_I + L_{II})/L_{III}$ suggest an M4 transition.

The decay of these conversion lines was followed, and both the K and L lines had a half-life of

$$T_{\frac{1}{2}\,(Bi^{201})} = 111 \pm 4 \text{ min},$$

as shown in Fig. 7, in good agreement with the value of 110 ± 20 min, found from our milking experiments, and with the value of 110 ± 10 min reported for Bi^{201} [31]. The only other bismuth isotope in this region which has a comparable half-life is Bi^{202}, and the only known isomeric state in lead which could follow the decay of the latter isotope is the 3.7-hr 9^- level in Pb^{202} [6, 13] which decays via a 787-keV E5 or a 129-keV E4 transition.

Thus we conclude that the correct assignment of this 629-keV M4 transition is to Pb^{201m} (61 sec) which follows the decay of Bi^{201} (111 min), and further, that this transition probably goes to the ground state of Pb^{201}.

Pb^{203m}

This isomer was observed following bombardments of lead with 40-, 50- and 70-MeV protons. Milking experiments were performed using the multichannel analyser

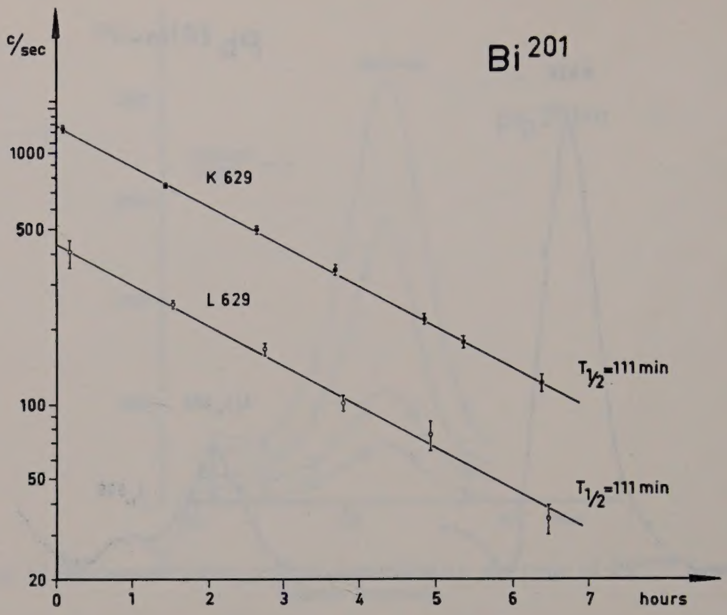

Fig. 7. Half-life of Bi^{201} obtained from the decay of the K and L conversion lines of the 629-keV gamma ray in Pb^{201m}.

in conjunction with the scintillation counter, and 5-sec counts were taken immediately after the milking and 3 minutes after the milking. The result of one experiment is shown in Fig. 8. The difference between the two spectra shows a strong peak at 830 ± 20 keV (mean value of five measurements is 825 ± 20 keV). The peak at 910 keV in the "background" spectrum is due to Pb^{204m}. By taking movie photographs of the scalers of the analyser channels in the region of this decaying peak, a half-life of about 6 seconds was found, the accuracy of this measurement being limited by the fact that the scalers of the multichannel instrument were scales of 10, so that it was not possible to take full advantage of the amount of activity which could be obtained in a single milking.

A more accurate half-life measurement was obtained with the same technique, but by using a single channel with a "window" of 100 keV centred on 825 keV and a scale of 100. The result is shown in Fig. 9, and gave a half-life of

$$T_{\frac{1}{2}\,(Pb^{203m})} = 6.5 \pm 0.5 \text{ sec.}$$

A further experiment was performed, in which the pulses from the scintillation counter were fed to each of two single channels, both of which were operated with a 100-keV window. One, the monitor, was centred on 825 keV, and the position of the other was varied. A series of milkings of the column was made, and each time the counts in 3-second intervals immediately after, and two minutes after the milking were measured in both the monitor and the variable channel. By taking the differences between these counts and by using a different position of the variable channel for each milking, the scintillation spectrum of the 6.5-sec activity was obtained. This

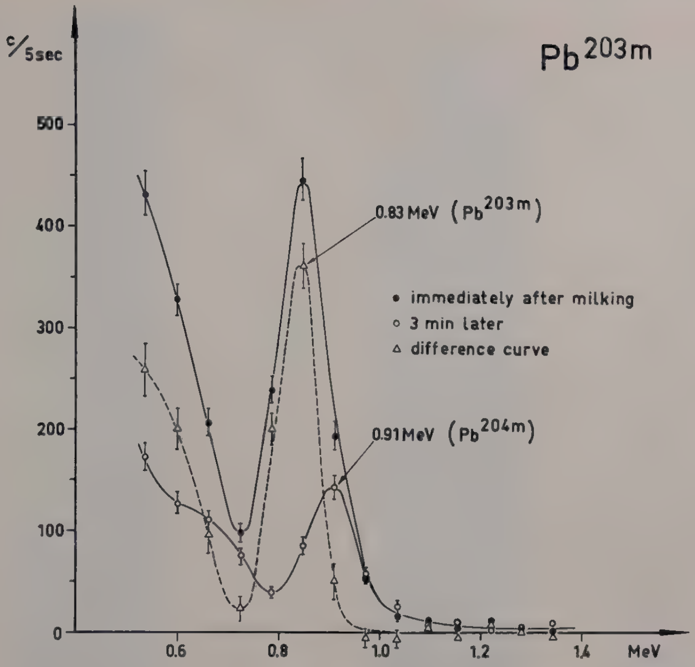

Fig. 8. Scintillation spectrum of 6.5-sec Pb^{203m} (dotted curve). With a multichannel analyser 5-sec counts were taken immediately after the milking (Pb^{203m} + 67.5-min Pb^{204m}) and 3 minutes after the milking (Pb^{204m}). The spectrum of Pb^{203m} was obtained by subtraction (the counting rate in the channel corresponding to $E > 1.43$ MeV was negligible).

is shown in Fig. 10, and it is fairly clear that no other gamma ray with energy larger than about 40 keV is in cascade with the 825-keV gamma ray. We have not had an opportunity to remeasure this spectrum with a greater precision in order to obtain the conversion coefficient of this gamma ray, but from Fig. 10 we can set a lower limit for the intensity of the X-ray peak, and after correction for crystal efficiencies and absorption in the glass, we find a lower limit for the K conversion coefficient

$$\alpha_K \sim 0.2.$$

This is to be compared with the theoretical conversion coefficients [27] of 0.22 and 0.036 for M4 and E4 transitions, respectively, of this energy.

Beta-spectrometer investigations of the corresponding bismuth activities have been made, using the double-focusing spectrometer of the Nobel Institute [25] and also a permanent magnet spectrometer for photographic recording [26]. As can be seen from the photographic film of Fig. 11 the conversion spectrum of Bi^{203} and Bi^{204} is very complex (cf. Appendix). The electron lines of a gamma ray with energy

$$E_{\gamma(Pb^{203m})} = 825.2 \pm 0.5 \text{ keV}$$

are very predominant. This gamma ray, converted in Pb, is with great certainty identical with the isomeric gamma ray found in the milking experiments.

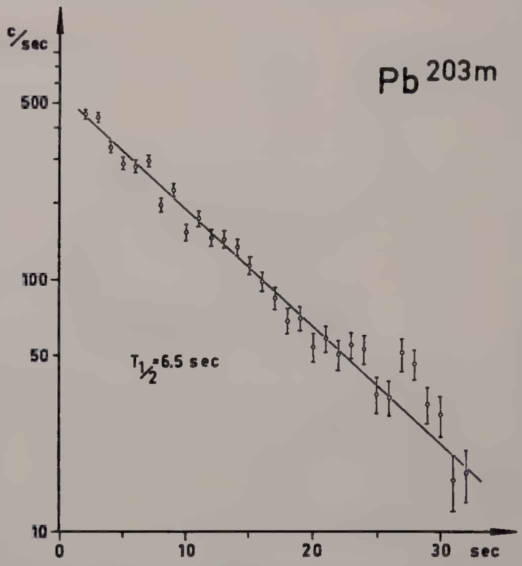

Fig. 9. Decay of Pb^{203m}. The half-life was obtained from movie photographs of the scaler of a single-channel analyser.

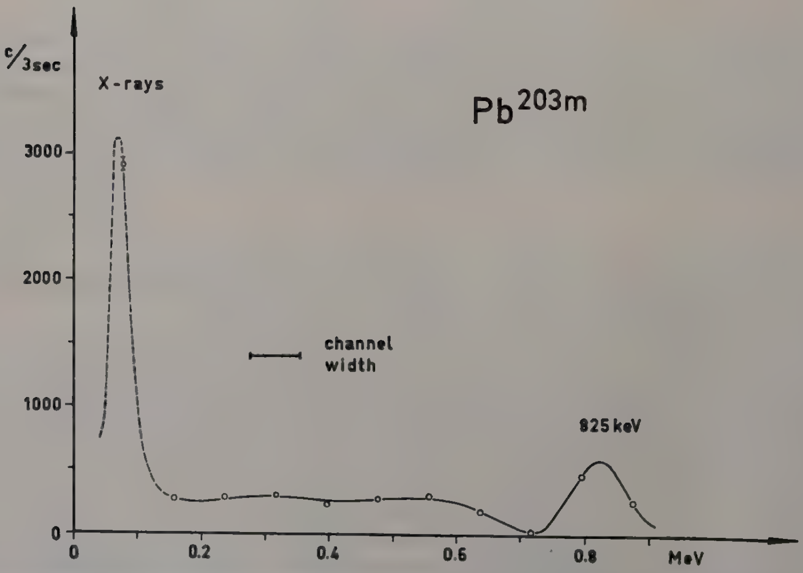

Fig. 10. Scintillation spectrum of Pb^{203m} obtained by a two-channel method described in the text.

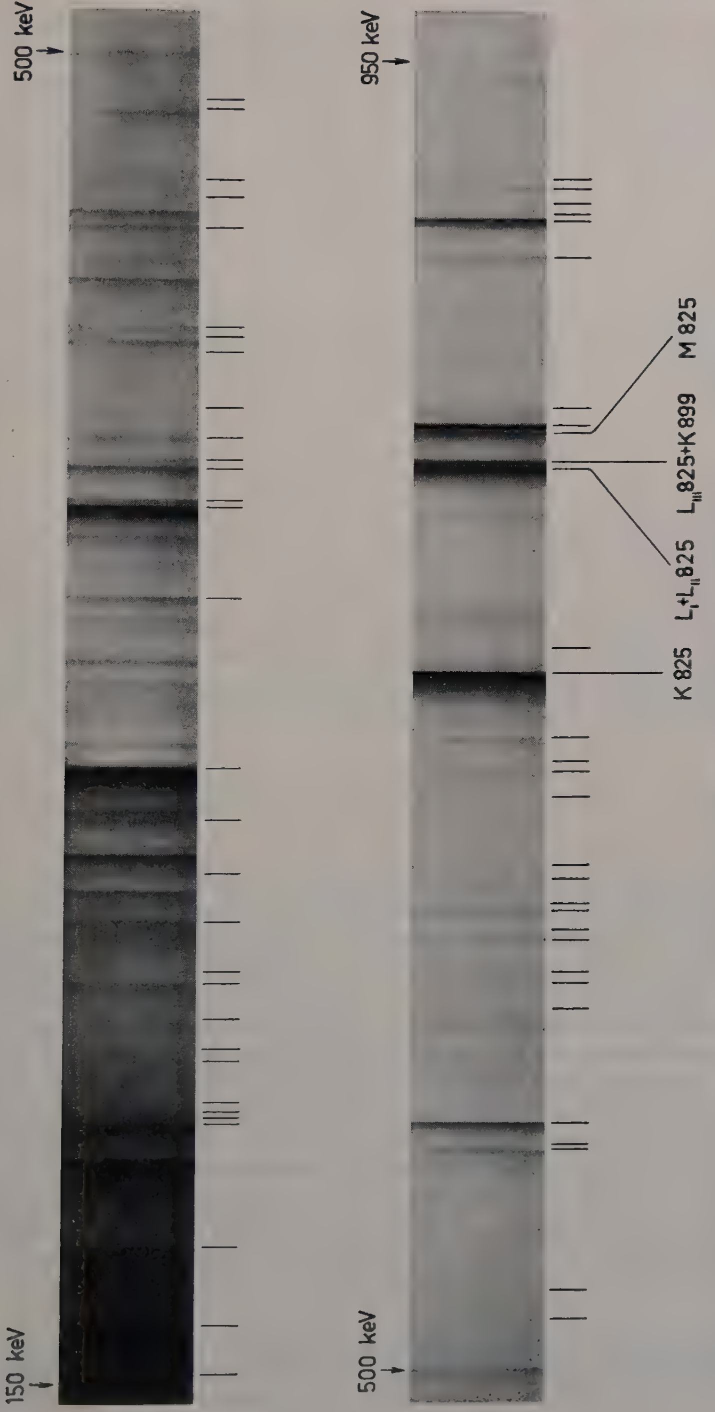

Fig. 11. β-ray spectrum of [Bi^{203} + Bi^{204}] (marked lines) + Bi^{205} + Bi^{206} recorded in a permanent magnet spectrometer. The conversion lines of the 825-keV gamma ray in Pb^{203m} are very pronounced. (Cf. Fig. 12.)

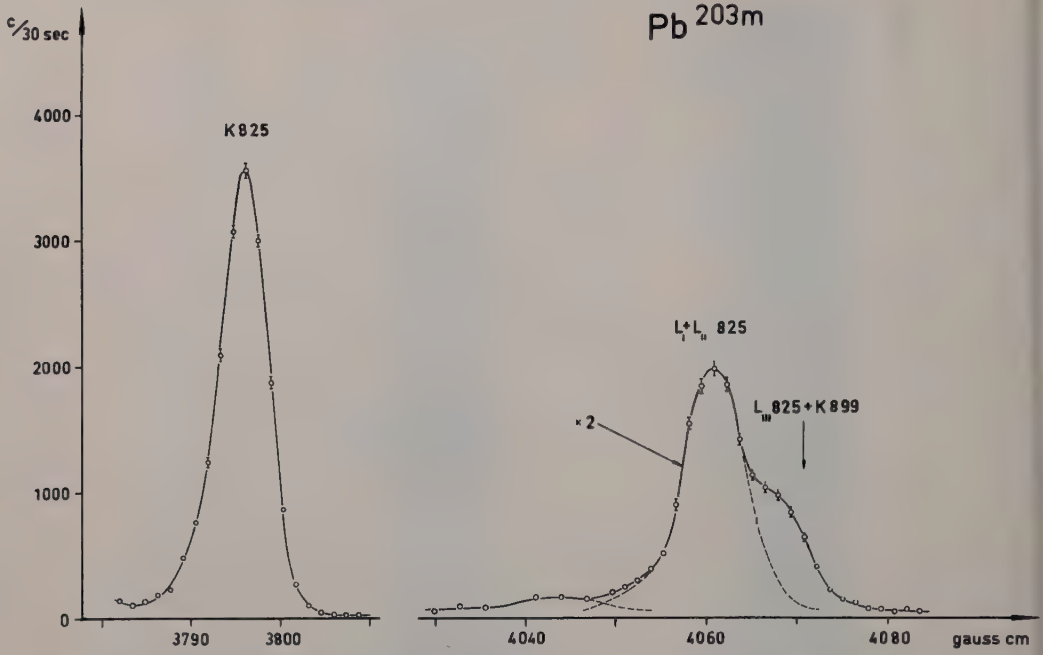

Fig. 12. Conversion lines of the 825-keV gamma ray in Pb^{203m} measured with 0.2 % resolution in a double-focusing β-spectrometer. The overlapping K 899 line is from Pb^{204m}. (Cf. Fig. 11.)

We obtained the value

$$T_{\frac{1}{2}\,(Bi^{203})} = 12.3 \pm 0.7 \text{ hr}$$

for the half-life of the bismuth parent, agreeing well with the reported value of 12 hr [11] for the half-life of Bi^{203}. The half-life of Bi^{204} is very nearly the same—13.5 ± 0.7 hr, according to our measurements—but the two isomeric levels which are known in Pb^{204} have half-lives of 67.5 min [17] and 0.26 μs [32]. Therefore we assign this 6.5-sec isomer to Pb^{203m}.[1]

A difficulty was encountered in measuring the K/L ratio of the 825-keV transition for the purpose of determining its multipolarity. In our bismuth sources the gamma rays from the decay of Bi^{204} are always present, and these include the transitions from the well-known 67.5-min level in Pb^{204m} to the Pb^{204} ground state. In this decay there are five gamma rays known, one of which has an energy of 899.3 keV [17]. Thus it is seen that the K conversion line of the 899.3 keV gamma ray has an energy which is only 0.9 keV smaller than that of the L_{III} line of the 825.2 keV transition. This corresponds to a momentum difference of 0.08 %, so that we are unable to resolve these lines in the double-focusing spectrometer, even under the best conditions. This part of the conversion spectrum is shown in Fig. 12 (cf. Fig. 11).

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[1] In a later experiment [50] Tl was bombarded with 38-MeV α-particles. In the conversion spectrum the 825-keV line was not observed at all, while the well-known lines of Pb^{204m} were predominant. This supports the assignment of the 825-keV line as a Pb^{203m} line.

However in this case also, the energy and half-life of the transition are such that the only reasonable assignments are M4 or E4. In either case the ratio $(L_I + L_{II})/L_{III}$ is large, being 9.1 for M4 and 12.5 for E4 [30]. Thus it should be possible to obtain an unambiguous assignment if the ratio $K/(L_I + L_{II})$ could be found, since the theoretical values of this ratio are 3.7 for M4 and 2.1 for E4 [30]. By assuming a reasonable shape for the high energy side of the $L_I + L_{II}$ line (dotted in Fig. 12), we obtained the ratio

$$K/(L_I + L_{II}) = 3.3 \pm 0.4,$$

indicating an M4 transition.

To sum up, therefore, we conclude that the 6.5-sec activity should be assigned to Pb^{203m} which follows the decay of 12.3-hr Bi^{203}, and that this isomer decays to the ground state of Pb^{203} via a single transition of M4 type, having an energy of 825.2 ± 0.5 keV.

Pb^{207m}

While we have observed this well-known 0.8-sec isomer, we have made no measurements which contribute anything new to what is already known about it. We mention its known properties here, because of their relevance to what follows. This isomer has been observed both in the decay of Bi^{207} [33] and following the reaction $Pb^{208}(\gamma, n)$ Pb^{207m} [22]. The most recent value of the half-life is 0.799 ± 0.013 sec [22], and in the isomeric decay there are two gamma rays in cascade, the level sequence being $i_{13/2} \rightarrow f_{5/2} \rightarrow p_{1/2}$ [34]. The energies of the two transitions are accurately known, that of the M4 transition being 1063.9 ± 0.3 keV [35] and that of the E2 transition 569.7 ± 0.2 keV [36].

Systematics of the odd-mass lead isomers

At this point we may conveniently discuss the systematic behaviour of certain features of the odd lead isomers dealt with above (see Table 1). In Fig. 13 the transition probabilities P of the isomeric transitions, divided by A^2 (A = mass number), are plotted as a function of their energies. $P = 1/\tau_\gamma$, where τ_γ, the mean life time of the gamma transition, is related to the observed half-life $T_{\frac{1}{2}}$ and the total conversion coefficient α by the expression $\tau_\gamma = T_{\frac{1}{2}}(1 + \alpha)/\ln 2$. It is seen that four of the experimental points lie very accurately on a straight line. The deviation of the fifth point (Pb^{197m}) from the line is due to the fact that no correction has been made for the electron capture branching that probably takes place from this isomeric level to a level in Tl^{197}. From this deviation a branching ratio $\lambda_{KC}/\lambda_{IT} = 4.1 \pm 0.5$ can be calculated.

In the same diagram are shown the corresponding curves for M4 transitions (with initial-state spin $I_i = 13/2$) as given by the semi-empirical formula of Goldhaber and Sunyar [29]

$$\tau_\gamma = 10^4 \frac{2 I_i + 1}{A^2 \cdot E^9} \cdot$$

and by Moszkowski [28]. According to Moszkowski P/A^2 is proportional to r_0^6 for M4 transitions (nuclear radius $r = r_0 \times A^{\frac{1}{3}}$). The curves are constructed for $r_0 = 1.45 \times$

Table 1. Isomeric transitions of M4 type in odd lead isotopes.

Isomer	Observed half-life	Transition energy (keV)	M4 assignment based on
Pb^{197m}	42 ± 2 [8] min	234 ± 0.5 [8]	K/L [8] $(L_I + L_{II})/L_{III}$ [8]
Pb^{199m}	12.2 ± 0.3 [10] min	424 ± 1 [8]	α_K * K/L [10] $(L_I + L_{II})/L_{III}$ [8] τ_γ *
Pb^{201m}	61 ± 2 * sec	629 ± 1 *	α_K * K/L * $(L_I + L_{II})/L_{III}$ * τ_γ *
Pb^{203m}	6.5 ± 0.5 * sec	825.2 ± 0.5 *	α_K * $K/(L_I + L_{II})$ * τ_γ *
Pb^{205m}	not observed		
Pb^{207m}	0.799 ± 0.013 [22] sec	1063.9 ± 0.3 [35]	α_K [37] τ_γ [29]

* This work.

10^{-13} cm [28], and for the most recent value $r_0 = 1.20 \times 10^{-13}$ cm [38], which gives better agreement with the experimental curve.

Fig. 14 shows the energies of the M4 transitions in Pb plotted as function of the neutron number of the isotopes in which they occur. A very smooth curve can be drawn through the points, and shows the characteristic rise in energy as the magic number is approached. This same tendency has already been seen in the corresponding curves for M4 isomers in Hg and Pt [3] (as well as for M4 transitions in nuclei with $N < 50$ and $N < 82$ [1, 2]), which are also shown in Fig. 14. Judging from the general systematic trend exhibited in Fig. 14, one would expect short-lived isomeric states also in Hg^{201}, Hg^{203} and Hg^{205}. Bergström *et al.* [39] tried to produce Hg^{201m} and Hg^{203m} in (γ, n) reactions, but no isomeric transition was found.

Shell theory suggests that all these M4 transitions in lead should take place between the $i_{13/2}$ and $f_{5/2}$ levels, as is known to be the case in Pb^{207m}. In all the odd lead isomers except Pb^{207m} only one gamma ray has been found, which leads to the conclusion that the ground states should be $f_{5/2}$. This is of interest in connection with the decay of the ground states. The corresponding thallium isotopes are expected to have the spin 1/2. The absence of K-capture transitions between the lead and thallium ground states favours an $f_{5/2}$ assignment of the lead ground states. Neither Prescott [40] nor

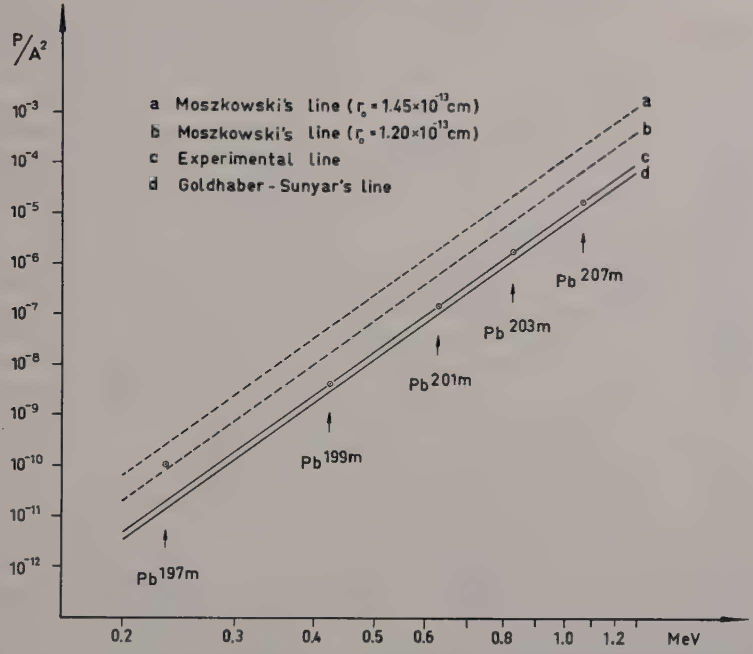

Fig. 13. Gamma decay-transition probability (P) divided by A^2 (A = mass number) as function of gamma-ray energy. (a) and (b) Theoretical curves for M4 transitions (with initial-state spin $I_i = 13/2$) according to Moszkowski (for $r_0 = 1.45 \times 10^{-13}$ cm and $r_0 = 1.20 \times 10^{-13}$ cm respectively). (c) Experimental points for odd lead isotopes. (d) Curve for M4 transitions (with $I_i = 13/2$) according to the semi-empirical formula by Goldhaber and Sunyar.

Wapstra *et al.* [41] observed any transitions between the ground states of Pb^{203} and Tl^{203}, in agreement with the level assignment above. The ground state of Hg^{203} has been assigned the spin $p_{3/2}$ or $p_{1/2}$, in spite of the fact that no beta transition has been observed between the ground states of Hg^{203} and Tl^{203}. The argument has been that no other odd-mass isomer of this type was known with the spin 5/2 in the ground state. Our experiments on the lead isomers have shown that this may be possible. The absence of an intense beta transition between the ground states of Hg^{203} and Tl^{203} may be explained by an $f_{5/2}$ ground state in Hg^{203}, and it is not necessary to assume some kind of "slowing-down mechanism" for allowed beta transitions in the neighbourhood of magic numbers [42]. A study of Hg^{203m}—if it exists—would clear up this question.

In all cases we have studied the lead isomers in equilibrium with their bismuth parents. The spin of the stable Bi^{209} is 9/2, in agreement with the shell theory prediction of an $h_{9/2}$ state for the 83rd proton [126 neutrons]. The ground state of Bi^{207} has also been assigned the spin 9/2 [43]. In order to decide if the ground states of Bi^{205}, Bi^{203}, Bi^{201} and Bi^{199} also have the spins 9/2, it is necessary to have more detailed information about the levels in the corresponding lead isotopes.

The notable omission in the curves of Figures 13 and 14 is the absence of points for the isomer Pb^{205m}. We shall later discuss the measurements we have made in an attempt to observe this isomer.

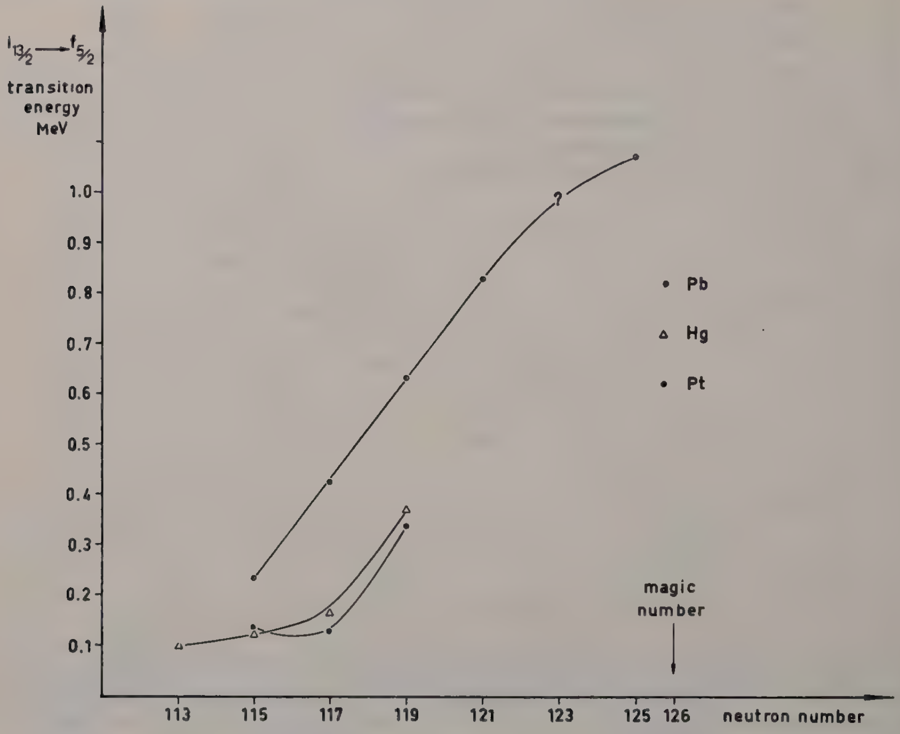

Fig. 14. Energies of $i_{13/2} \rightarrow f_{5/2}$ transitions in Pb, Hg and Pt as functions of neutron number. As indicated in the figure, no isomeric transition was observed in Pb^{205} ($N = 123$).

Matrix elements of the 13/2+ → 5/2− transitions

The radial matrix elements for an M4 transition between two nuclear states i and f may be written [28]:

$$|M|^2 = \int \psi_i(r)\, H'\, \psi_f(r)\, dV$$
$$= 1.16 \cdot 10^6 \cdot r_0{}^6 \cdot A^{-2} \cdot E_\gamma^{-9} \cdot S^{\;1} \cdot T_{\frac{1}{2}}^{-1} \cdot \left[1 + \alpha_K \left(1 + \frac{\alpha_L + \alpha_M + \cdots}{\alpha_K}\right)\right]^{-1},$$

where

$r_0 = 1.20$ [38]
A = mass number
E_γ = energy of the M4 γ-ray (in MeV)
S = statistical factor [28]
$T_{\frac{1}{2}}$ = observed half-life of the isomeric transition
α_K, α_L, $\alpha_M \cdots$ = K, L, M ⋯ conversion coefficients.

The statistical factor S depends on the spin values of the states i and f. For 13/2+ → 5/2− transitions the value of S is 225/143. It should be pointed out that there is no definite experimental evidence that the M4 gamma-rays in the odd lead isotopes are due to 13/2+→5/2− transitions, although this is suggested by the shell model.

The value of $|M|^2$ can be calculated from the experimentally determined values of A, E, $T_{\frac{1}{2}}$ and α_K, α_L, α_M ... However, the energy in particular must be measured very accurately because of the E_γ^9-dependance. Furthermore, in many cases theoretical K conversion coefficients must be used. It has recently been shown by L. Sliv *et al.* [27] that the previously used K conversion coefficients of M. E. Rose *et al.* [30] are about 15 % too large for M4 transitions in the Pb region. Hence the Rose values give too small $|M|^2$ values, the deviation being larger for the more neutron deficient isotopes (small E_γ, large α_K). This introduces an apparent increase of $|M|^2$, when N approaches the magic number 126. Table 2 shows $|M|^2$ for the M4 transitions in Pb, based on the most accurate values of E_γ, $T_{\frac{1}{2}}$, $(\alpha_L+\alpha_M+\ldots)/\alpha_K$* and the theoretical K conversion coefficients of L. Sliv *et al.* [27].

Table 2. Values of $|M|^2$ for M4 transitions in lead**

N	117	119	121	123	125
$\|M\|^2$	3.7±0.2	3.5±0.2	4.0±0.4		3.7±0.1

It has been pointed out by several authors [3, 7, 28] that the matrix elements for M4 transitions increase close to magic numbers. As can be seen from Table 2 there is no increase (within the experimental errors) of $|M|^2$ towards N = 126 in the case of Pb. Previous conclusions [3, 7] that such a trend existed for the Pb isomers, appear to be due to inaccurate energy determinations and the use of too high theoretical K conversion coefficients.

The increase of $|M|^2$ has been explained by the argument [28] that excited states of nuclei should be represented by relatively "purer" particle wave-functions, when the neutron number approaches a magic number. At first sight our results seem to contradict this argument. In this case, however, there might be additional effects which might cancel out the increase of $|M|^2$ close to the magic number 126: the excitation energy of the isomeric state in Pb^{207} is so high that admixture of core-excited states is to be expected. This means a new kind of admixture, which might compensate for the decrease of admixture due to the decreasing number of combinations of particle states when approaching the magic number.

Search for Pb^{205m}

Bi^{205} (14.5 d) [11] was produced by the reaction Pb^{206} (d, 3n) in bombardments of natural lead with 25 MeV deutrons in the Nobel Institute cyclotron. In such bombardments the isotopes Bi^{206} (6.4 d) [11] and Bi^{207} (27 y) [21] are also formed. From the curves of Figures 13 and 14, one would expect an M4 transition in Pb^{205} to have an energy of 980 ± 40 keV, and a half-life of 1.5 ± 0.5 sec.

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* $(\alpha_L+\alpha_M+\ldots)/\alpha_K \sim \frac{4/3}{\alpha_K/\alpha_L}$. The α_K/α_L ratios were calculated by use of Sliv's K conversion coefficients and Rose's L conversion coefficients with α_{L_I} corrected (for nuclear size) by the same relative amount as α_K. (However, when Rose's ratios were used instead, the figures in Table 2 remained unchanged.)

** $|M|^2 = 3.8 \pm 0.5$ for N = 115 [8].

Milking experiments were performed with our standard technique, and the scintillation spectrum of the milked activity observed with the multichannel analyser. The differences between the counts in 3-second intervals immediately after and 2 minutes after each milking were added for several milkings to give the counts in each channel due to any short-lived activity. The result of the series of measurements in which the best statistics were accumulated is shown in Fig. 15. The curve in the figure is the spectrum of Pb^{207m}, obtained by taking the spectrum of a Bi^{207} source, and correcting for the fact that only 87 % of the decays of Bi^{207} feed the 1064 keV gamma ray, while all decays feed the 570 keV gamma ray [21].

It is clear that the spectrum we have thus obtained is essentially that of the Pb^{207m} milked out of the Bi^{207} in our source. There is only one anomalous point, that in channel 7 ($\sim$ 730 keV), which could be taken to suggest the presence of any other activity, but the curve of Fig. 14 shows that it is most unlikely that any M4 transition of such an energy would exist in this isotope. Since we have, in another investigation [18] which is as yet uncompleted, been studying the decay of Bi^{205}, we can say something about the gamma rays which could be associated with any possible isomer in Pb^{205} (see Appendix).

Firstly, there is in fact a strong gamma-ray of energy 703 keV, as has already been reported by Alburger and Pryce [44]. However, we have measured its K/L ratio as 4.2 ± 0.3 in the double-focusing spectrometer and its K conversion coefficient as 0.009 ± 0.003 in an e^-e^--coincidence experiment [18]. These results indicate an E2 transition (cf. $K/L = 3.9$ [30] and $\alpha_K = 0.010$ [27]) and certainly exclude the possibility of M4 transition (cf. $K/L = 2.9$ [30] and $\alpha_K = 0.39$ [27]).

Secondly, there is a fairly strong gamma ray of 988 keV. It is difficult to measure its K/L ratio since its L line lies very close to, and on the low energy side of the strong Bi^{207} 1064 K line which always is present in our spectra. Our best estimate of this K/L ratio is 4.2 ± 0.8, and on this basis an M4 assignment may be possible (cf. $K/L = 3.7$) [30]. However, this possibility can be rejected on the following grounds. From the bismuth activity used in the milking experiment which gave the results of Fig. 15, a source was prepared for the double-focusing spectrometer. In a measurement taken 16 days after the milking experiment, the intensity of the 988 K Bi^{205} line was 0.5 times that of the 1064 K Bi^{207} line. Thus, if the 988-keV transition were of M4 type, its gamma ray should have been about as strong as the 1064-keV gamma ray at the time of the milking experiment, and its half-life would be, as mentioned earlier, about 2 sec. There is clearly no evidence for such a gamma ray in the spectrum of Fig. 15.

For this same reason it is fairly sure that the gamma rays of 1003 keV, 1014 keV and 1044 keV, none of which is considerably stronger than the 988-keV gamma ray (see Appendix), cannot be M4 transitions. Lastly, there is no other gamma ray observed in the decay of Bi^{205} whose energy could fit the curve of Fig. 14.

Thus we are forced to the conclusion that if the $i_{13/2}$ and $f_{5/2}$ levels occur in the level scheme of Pb^{205}, it is probable that no transition between them takes place with observable intensity. Evidence supporting this view comes from the above-mentioned investigation of Pb^{207m} by Bendel, Toms and Tobin [22]. Since this isomer was produced by the reaction Pb^{208} (γ, n) in a target of natural lead, one would have expected them to observe Pb^{205m}, if it existed, from the reaction Pb^{206} (γ, n). Their decay curve shows what appears to be a pure 0.8-sec activity, and Dr. Bendel has

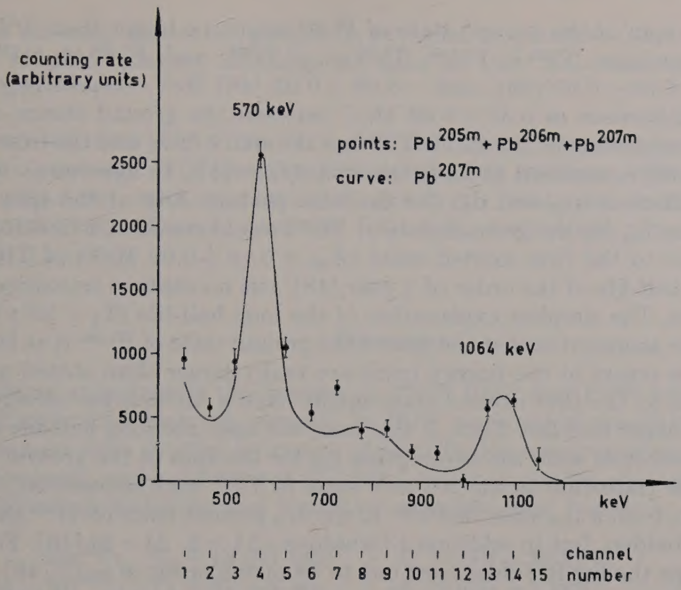

Fig. 15. Scintillation spectrum (points) obtained with a multichannel analyser in a search for Pb^{205m} (counts accumulated from 11 milkings). For comparison, the figure includes the spectrum of Pb^{207m} (curve), obtained as described in the text and arbitrarily fitted to the experimental point in channel 4.

informed us that, on the assumption that Pb^{205m} exists and is of the same type of isomer as Pb^{207m}, the $Pb^{206}(\gamma, n)\,Pb^{205m}$ cross-section for 20-MeV bremsstrahlung is less than about 20 % of that for $Pb^{208}(\gamma, n)\,Pb^{207m}$.

The failure of observing M4 radiation emitted from Pb^{205m} might be explained in several ways, the most plausible of which are discussed below:

1. The spin or decay energy of the 14.5-day state in Bi^{205} might be such that the probability of electron capture to the $i_{13/2}$ state or higher states in Pb^{205} would be very small. However, this explanation does not account for the negative result of the (γ, n) reaction [22].

2. Knowing the levels of Pb^{207} (one hole in the "$N = 126$ shell") Alburger and Pryce [44] could successfully predict several levels in Pb^{206} (two holes). A use of the same method in order to predict the levels of Pb^{205} (three holes) would result in a much more complicated level scheme than that of Pb^{207}. In fact the level scheme of Pb^{205} (more than 30 gamma rays, see Appendix) is much more complicated than that of Pb^{207} (5 gamma rays) [21]. From this point of view it would not be surprising if there were a level situated between the 13/2 and 5/2 levels having an intermediate spin and thus making an observation of M4 radiation from the 13/2 level impossible. On the other hand, it is not at all clear why the isomeric transition should "reappear" in Pb^{203}, Pb^{201}, Pb^{199} and Pb^{197}.[1]

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[1] Dr. K. Bleuler, Zürich, suggests that a complex state of intermediate spin is situated between the $i_{13/2}$ and $f_{5/2}$ levels in Pb^{205} and thus destroys the metastability of the $i_{13/2}$ level. In Pb^{203} the $i_{13/2} \rightarrow f_{5/2}$ energy difference is smaller than in Pb^{205} (see Fig. 14), so that this intermediate spin state might cross the $i_{13/2}$ level. In general, the excitation energies of one-particle (-hole) states decrease when receding from a magic number (due to pairing energy), while the energies of complex states might remain at the same value.

3. Finally, the spin of the ground state of Pb^{205} might be larger than 5/2. The Q values of the reactions Tl^{206} $\xrightarrow{\beta^-}$ Pb^{206}, Tl^{205} (n, γ) Tl^{206}, and Pb^{206} (d, t) Pb^{205} are 1.51 ± 0.01 [45], 6.20 ± 0.03 [46], and -8.09 ± 0.05 [46] MeV, respectively. These give an energy difference of 0.38 ± 0.09 MeV between the ground states of Pb^{205} and Tl^{205}. The ground state of the stable Tl^{205} has the spin 1/2 +, and the first excited state (205 keV) [46] is assumed to have the spin 3/2 + [47], in agreement with the shell theory predictions $s_{1/2}$ and $d_{3/2}$ for the 81st proton. Any of the spin assignments $p_{1/2}$, $p_{3/2}$, or $f_{5/2}$ for the ground state of Pb^{205} would result in a first forbidden capture transition to the first excited state ($E_{\mathrm{EC}} = 0.18 \pm 0.09$ MeV) of Tl^{205}. This corresponds to a half-life of the order of 1 year [48], but no capture transition has as yet been observed. The simplest explanation of the long half-life ($T_{\frac{1}{2}} > 10^6$ y) [19] of Pb^{205} would be the assumption that the spin of the ground state of Pb^{205} is at least 7/2.

However, if the errors in the energy cycle are really larger than stated above, a capture transition to the first excited state might have a much lower energy and a correspondingly longer half-life. Thus, if this were the case, the long half-life of Pb^{205} might be explained even with the assumption $f_{5/2}$ for the spin of the ground state of Pb^{205}. If a capture transition to the 205-keV state in Tl^{205} were energetically impossible, a transition from a $f_{5/2}$ state of Pb^{205} to the $s_{1/2}$ ground state of Tl^{205} should be not only first forbidden, but in addition l-forbidden ($\Delta I = 2$, $\Delta l = 3$) [49]. For other decays of this type the log ft-values turn out to be of the order of 8–12 [49]. Such a value would not be completely incompatible with the long half-life of Pb^{205}.

De-Shalit and Goldhaber [42] assumed that the long half-life was due to the slowness of the transition

$$\underbrace{(s_{1/2})^2\,(h_{11/2})^{12}}_{\text{protons}}\;\underbrace{(i_{13/2})^{12}\,(p_{1/2})^1}_{\text{neutrons}} \longrightarrow \underbrace{(s_{1/2})^1\,(h_{11/2})^{12}}_{\text{protons}}\;\underbrace{(i_{13/2})^{14}\,(p_{1/2})^0}_{\text{neutrons}}$$

which does not only involve a single-particle transition but also a change of the core configuration.

It is evident from the discussion above that it is impossible to make any definite conclusions about the failure of observing Pb^{205m}, before our investigation of the decay of Bi^{205} is completed. However, already at this stage we can exclude the possibility of the spin value 1/2 for the ground state of Pb^{205}, assuming that the ground state of Bi^{205} is $h_{9/2}$.

Appendix. Investigations of Bi^{203}, Bi^{204} and Bi^{205}

1. The following gamma-ray energies (in keV) and K conversion-line intensities (w = weak, vs = very strong, etc.) were found in the decay of Bi^{203} (12.3 h), (conversion in Pb), [50]:

186.5 (w), 263.2 (s), 721.8 (w), 825.2 (vs).

2. The following gamma-ray energies (in keV) and K conversion-line intensities were found in the decay of Bi^{204} (13.5 h), (conversion in Pb), [50]:

91.2 (w), 95.3 (w), 140.8 (vw), 218.9 (w), 240.9 (w), 248.9 (s), 288.0 (s), 374.7 (vs), 412.5 (vw) 422.0 (w), 501.8 (w), 522.6 (w), 533.3 (s), 660.9 (w), 662.0 (vw), 670.8 (s), 709.7 (vw), 718.4 (vw), 725.1 (vw), 898.8 (vs), 911.8 (vs), 917.7 (vw), 1.8×10^3.

3. The following gamma-ray energies (in keV) and, in parenthesis, K-conversion-line intensities (K 703.3-line intensity arbitrarily set equal to 1000) were found in the decay of Bi^{205} (14.5 d), [18]:

115.2 (100)[1], 235.9 (80), 260.5 (1100), 282.3 (410), 284.2 (1600), 349.4 (280), 383.2 (60), 493.6 (110), 511.7 (220), 550.0 (63), 571.0 (740), 580.0 (240), 626.5 (67), 688.9 (10), 703.3 (1000), 745.0 (21), 758.3 (31), 761.0 (20), 910.8 (98), 987.8 (275), 1002.7 (21), 1014.2 (49), 1043.7 (325), 1190.3 (76), 1337 (3), 1351.5 (11), 1502.5 (4), 1552.0 (8), 1614.6 (31), 1766.4 (436), 1777.4 (52), 1863.3 (20), 1906.5 (6), 2.6×10^3.

Summary

In milking experiments the isomers of Pb^{199}, Pb^{201} and Pb^{203} were obtained from their corresponding bismuth parents. In Pb^{205} no isomeric transition was observed.

The isomers were studied by means of scintillation spectrometers, double-focusing β-spectrometers, and a permanent magnet β-spectrometer for photographic recording.

The following half-lives and transition energies were found:

Pb^{199m}: $T_{\frac{1}{2}} = 13 \pm 1$ min, $E_\gamma = 410 \pm 30$ keV;
Pb^{201m}: $T_{\frac{1}{2}} = 61 \pm 2$ sec, $E_\gamma = 629 \pm 1$ keV;
Pb^{203m}: $T_{\frac{1}{2}} = 6.5 \pm 0.5$ sec, $E_\gamma = 825.2 \pm 0.5$ keV.

The isomeric transitions were shown to be M4 transitions. Only one gamma ray was found in each decay. Therefore we suggest $f_{5/2}$ ground states for Pb^{199}, Pb^{201} and Pb^{203}.

Odd lead isomers were shown to exhibit a systematic trend in half-lives and energies.

The matrix elements of the M4 transitions in the odd lead isomers were found to be remarkably constant.

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[1] L-line intensity.

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